Structural Study of Al-doped ZnO Thin Films Produced by the Sol-gel Technique

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Keywords: thin films, spin coater, ZnO / Al, XRD, SEM, UV-Visible

Abstract: In this work we studied the effect of aluminum doping concentration on the structural properties of zinc oxide thin films, we deposited samples of ZnO and aluminum doped ZnO with a doping rate of 1, 2, 3, 4 and 5%, on glass substrates by the spin coating technique. The structural characterization of the samples is done by the XRD and SEM techniques, the XRD spectra show that the layers are polycrystalline with a hexagonal würtzite structure, and a preferred orientation in the plane (002), and for the doping 5% the structure is almost monocrystaline (002). SEM images are used to confirm grain sizes and surface conditions. Optical characterization is done by UV-visible spectroscopy, gives a good visible transmittance up to 80% and exceeds 90% for 2% doping, and the gap varies with the doping variation with a small gap for the same doping (2%).

1 INTRODUCTION

In recent years, transparent and conductive oxides (TCO) such as ZnO, find important applications in new technologies. Since the discovery at the beginning of the century of duality: good electrical conductivity and good optical transparency in the visible, research in the field of TCO are really intensified from the 80s. Non-toxicity and this great existence on earth are the main advantages of ZnO, these advantages give the possibility of reducing the manufacturing costs of ZnO-based composites, in the massive state the ZnO has several structures, among its structures, the Würtzite structure is the most stable for ZnO, these lattice parameters of the latter are: a = 0.3296 nm and c = 0.5207 nm. It does not have a center of symmetry. It can be deduced that this structure is an insertion of two HC networks (the O2- and Zn2 + network), with the displacement of the oxygen network by a fraction of 0.38 the size of a unit cell relative to ZnO is a degenerate semiconductor with n-type conductivity, this conductivity is due in particular to excess zinc in interstitial positions, with a large direct gap (3.436 to 0 K and 3.2 eV at room temperature).). Its electrical properties can be changed by heat treatment, or by appropriate doping, the conductivity of ZnO can be increased by the substitution of trivalent atoms for the positions of the Zn atoms, and to reduce the conductivity the Zn atoms can be substituted by monovalent atoms.

The methods of elaboration of the thin layers are classified in two big categories: Physical methods (Sputtering, Laser ablation, Evaporation under vacuum), and Chemical methods (Spray pyrolysis, Sol gel), in this work we prepared our layers by the Sol-Gel method associated with spin coater.

2 EXPERIMENTAL PART

The ZnO layers are deposited based on a solution that prepares as follows: zinc acetate dihydrate [Zn (CH_3COO) 2. 2H2O] (a concentration of 0.4M / L) dissolved in a mixture of isopropanol, ethanol and monoethanolamine, this mixture and heated stirring with a temperature of 60 ° C for a period of 2 hours. The dopant source (Aluminum) is aluminum chloride ($AlCl_3$). The deposit is made by

Boufelgha, F., Brihi, N., Bouaine, A., Zellagui, R., Ouafak, N. and Boughelout, A.
Structural Study of Al-doped ZnO Thin Films Produced by the Sol-gel Technique.
DOI: 10.5220/0009772001290132
In *Proceedings of the 1st International Conference of Computer Science and Renewable Energies (ICCSRE 2018)*, pages 129-132
ISBN: 978-989-758-431-2

the spin-coating technique (rotation of about 2500 rpm) for 30s, and drying between each 2 layers at 200 ° C for 10 minutes, and a final annealing at 600 ° C for 3 hours.

We can schematize the steps followed for the preparation of our samples as follows:

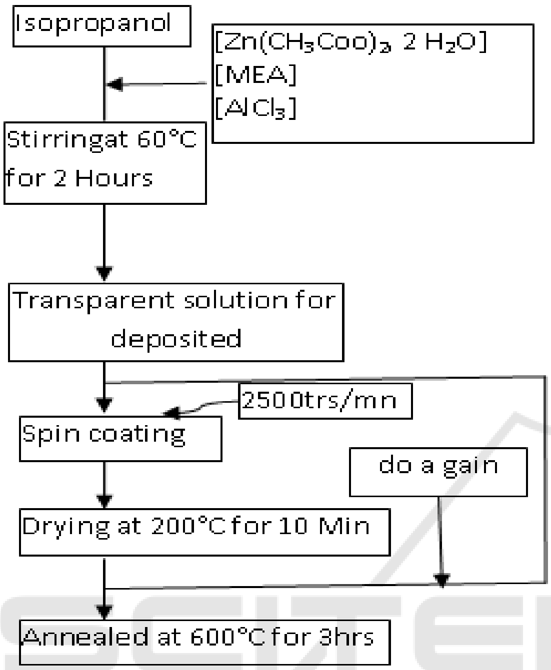

Fig.1 Schematization of the steps followed for the elaboration of our samples.

3 RESULTS AND DISCUSSIONS

3.1 Structural Properties

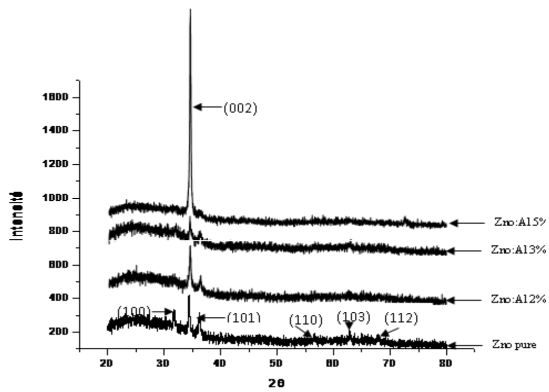

Fig.2 ZnO DRX Spectra: Pure, Doped Aluminum 2, 3 and 5%

Fig.2 shows the X-ray spectra of thin layers of zinc oxide, deposited by the sol-gel method (spin-coating technique), pure samples, doped with aluminum with different concentrations 2, 3 and 5%. The identification of the peaks is made by comparing them with the diffraction angles found on ASTM sheets 36-1451 (American Society for Testing and Materials). The spectra affirm a crystallization. For the ZnO which one can not speak about a growth in a preferential direction (the appearance of 3 peaks of the same intensity) almost: (100), (002) and (101), in the case of ZnO doped on speaks of growth in a preferential orientation according to the direction (002). One can speak to almost total growth in the (002) direction for the 5% concentration. These results confirm the results found by F. Chouikh for pure and Al doped ZnO samples deposited by the pyrolysis spray technique.

Texture

For the purpose of checking the preferred orientation, the ratio R_{xi} which is given by: $R_{xi} = I_{xi} \sum_{n=0}^{N} I_{xn}$

For pure ZnO: there are 5 peaks with the concentrations: $I_{(100)}$=283, $I_{(002)}$=450, $I_{(101)}$=251, $I_{(103)}$=258, $I_{(112)}$=164,

$R_{(100)}= I_{(100)} /[I_{(100)}+ I_{(002)}+ I_{(101)}+ I_{(103)}+ I_{(112)}]$=283/1306=0.217

$R_{(002)}= I_{(002)} /[I_{(100)}+ I_{(002)}+ I_{(101)}+ I_{(103)}+ I_{(112)}]$=450/1306=0.344

$R_{(101)}= I_{(101)} /[I_{(100)}+ I_{(002)}+ I_{(101)}+ I_{(103)}+ I_{(112)}]$=251/1306=0.192

$R_{(103)}= I_{(103)}/[I_{(100)}+ I_{(002)}+ I_{(101)}+ I_{(103)}+ I_{(112)}]$=158/1306=0.121

$R_{(112)}= I_{(112)} /[I_{(100)}+ I_{(002)}+ I_{(101)}+ I_{(103)}+ I_{(112)}]$=164/1306=0.126

in the same way, we find:

Table 1: Texturing variation of orientations for each doping rate.

	(100)	(002)	(101)	(103)	(112)
0	0.217	0.344	0.192	0.121	0.126
2	0.000	0.832	0.088	0.080	0.000
3	0.000	0.847	0.072	0.081	0.000
5	0.000	0.850	0.087	0.000	0.063

For the undopedZnO, the growth does not have a preferential direction, we notice the appearance of the peaks (lines): (100), (002), (101), (103) and (112) with almost the same concentrations (intensities), this non-texturing is similar to the surface energies of the three lines.

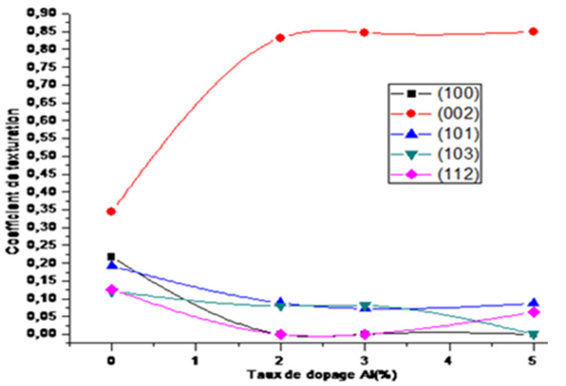

Fig.3 Variation of texture coefficients according to doping rates.

In the case of doped ZnO, an increase in the texture coefficient of the line (002) as a function of ZnO doping rate with Al is observed, a maximum texturing (R (002)) is observed on the line (002) for the concentration 5%.

3.2 Morphology of the Layers

Fig 4, 5 and 6 show the SEM images of aluminum doped ZnO samples by percentages 2%, 3% and 5% respectively.

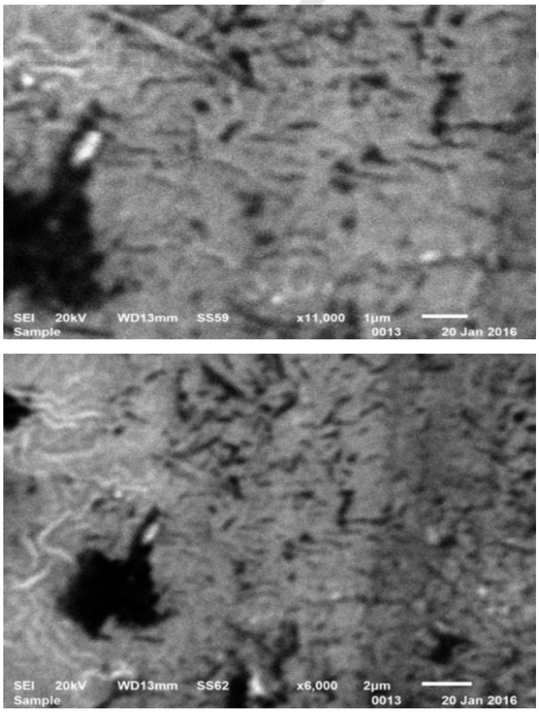

Fig.4 ZnO/Al 2%

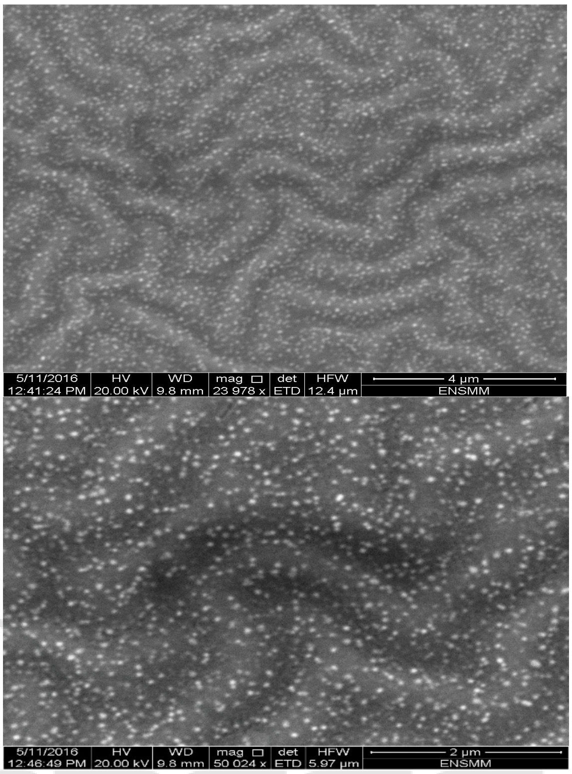

Fig.5 ZnO/Al 3%

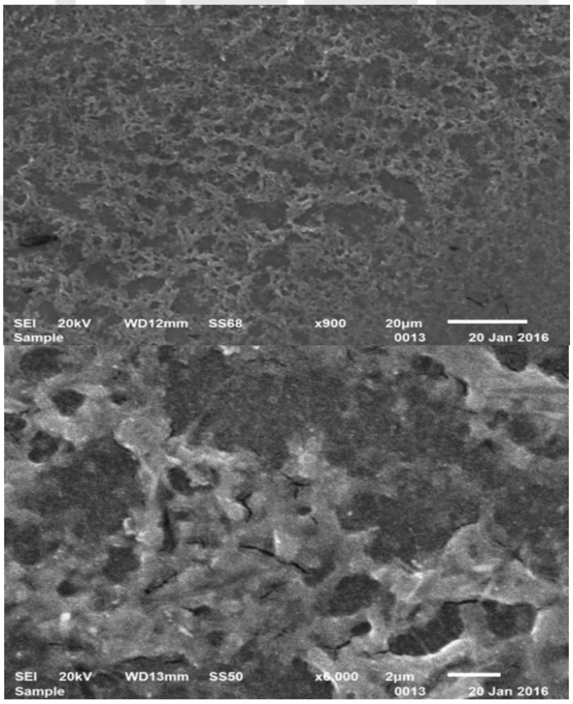

Fig.6 ZnO/Al 5%

The SEM images show a homogeneity of the morphology of the surface of all the samples.

Fig.4 (2% doping) has a homogeneous can with the presence of rod-shaped relief.

Fig.6 (5% doping) has a melted and porous layer showing the presence of some microcracks at the surface.

For Fig.5 (3% doping) has a completely homogeneous layer, with the presence of shape grains.

3.3 Optical Properties

Fig.7 represents the UV-visible spectra of aluminum doped ZnO samples (Al) in percentages 1, 2, 3, 4 and 5%.

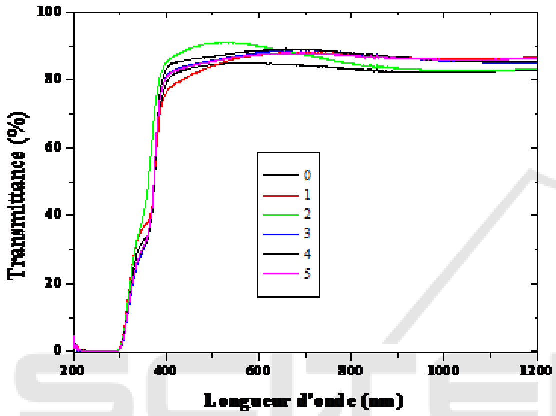

Fig.7 Transmittance spectra of ZnO and ZnO / Al (1, 2, 3, 4 and 5%)

The UV spectra show a transmittance higher than 80% in the visible for all the samples, the transmittance exceeds the 90% for the 2% doping, one can hard that the latter to a better transmittance compared to the others.

Fig.8 shows the optical Gaps of ZnO and ZnO / Al (1, 2, 3, 4 and 5%).

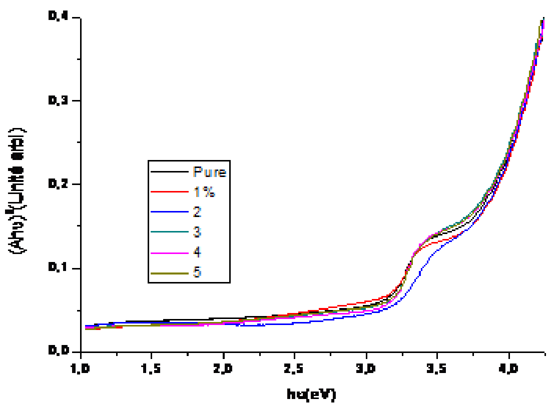

Fig.8 ZnO and ZnO / Al Optical Gap (1, 2, 3, 4 and 5%)

Fig.9 shows the variation of the ZnO / Al gaps

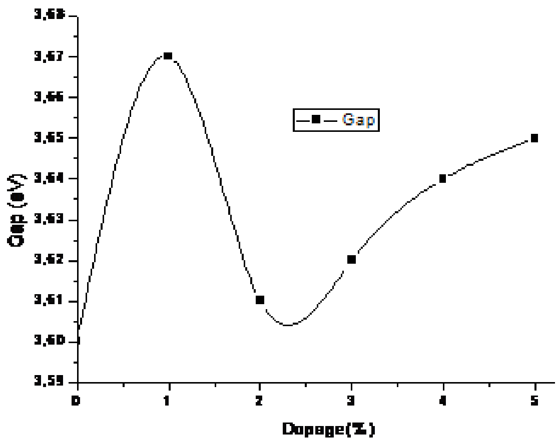

Fig.9 optical Gap variation of ZnO and ZnO / Al (1, 2, 3, 4 and 5%)

4. CONCLUSION

The thin layers of undoped and Al doped ZnO were developed by spin-coater-associated sol-gel methods on glass substrates. Aluminum doping changes the properties of our layers (structural and optical). the results of the XRD show a preferential orientation according to the plan (002). The UV-Visible results show a transmittance of over 80%. Note for concentration 5% the crystal and almost mono.

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